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LETTER TO THE EDITOR

Effect of noise on spinodal decomposition

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Abstract. The long-time behaviour of two-dimensional systems undergoing spinodal decomposition is studied numerically with the aid of a cell-dynamical approach both without and with noise. In both cases, the representative length scale of the pattern behaves as $l(t) \sim t^{\phi}$, where the exponent ϕ crosses over from ~ 0.28 to ~ 0.33. The crossover time increases with an increase in amplitude of the noise.

An outstanding problem in studying the kinetics of phase transitions is that of the mechanisms whereby ordering takes place in thermodynamically unstable systems, e.g. quenched binary alloys (Gunton *et al* 1983, Lebowitz *et al* 1983, Furukawa 1985). The ordering process depends critically on whether or not there is a conserved order parameter. In this letter we numerically demonstrate, using a cell-dynamical system approach, that there is a crossover in the domain growth law for the conserved case from $l(t) \sim t^{0.28}$ to $l(t) \sim t^{0.33}$, where $l(t)$ is the typical domain size at time t. Furthermore, we demonstrate the existence of a 'scaling regime' (explained later) in which the effect of noise appears to be unimportant. The results presented here are for two-dimensional systems with critical quenching, i.e. the spatial integral of the order parameter is zero for all times. This is an example of what has been termed 'spinodal decomposition'.

The theoretical study of phase separation in the conserved case dates back to the classic work of Cahn and Hilliard (1958) and the work of Cook (1970), who considered a (phenomenological) partial differential equation model, now called the Cahn-Hilliard-Cook (CHC) equation.

Interest in the study of spinodal decomposition was revitalised by the observation of an approximate scaling law in Monte Carlo simulations of the process (Marro *et al* 1979, Lebowitz *et al* 1982, Sahni and Gunton 1980). These suggested that the normalised form factor $S(\boldsymbol{k}, t)$ has a scaling regime in which it behaves as

$$S(\boldsymbol{k}, t) = l(t)^d \Phi(\boldsymbol{k} l(t)) \tag{1}$$

where $\boldsymbol{k}$ is the wavevector, t the time, Φ a universal function, $l(t)$ a time-dependent length scale which behaves as $l(t) \sim t^{\phi}$ for some positive number ϕ and d is the spatial dimensionality. (It should be noted that, previous to these results, Binder and Stauffer (1974) and Furukawa (1977, 1978, 1979) had discussed possible scaling of the form factor.) Marro *et al* (1979) performed extensive Monte Carlo simulations using a spin-exchange kinetic Ising model. They found that ϕ varies between 0.19 and 0.3 depending on the depth of quenching, the off-criticality and the time of evolution.

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There is an approximate scaling in the sense that data for fairly long periods of time can be fitted to master curves. However, the shape of the master curve appears to change gradually in time so that data for very long time periods cannot be fitted to a single curve. Huse's Monte Carlo results (Huse 1986) suggest (by extrapolation) a late-stage domain growth law of $l(t) \sim t^{0.33}$.

The CHC equation has also been the subject of considerable numerical work. It was solved by Petschek and Metiu (1985) and (without noise) by Miyazaki *et al* (1983). However, they did not study the scaling regime or the domain growth law. Recently, Gawlinski *et al* (1987) reported a numerical study of the CHC equation. They found a power-law behaviour with $\phi \sim 0.33$.

Given the numerical effort involved in successfully simulating the CHC equation or performing a Monte Carlo simulation, it is desirable to have computationally efficient models to study the scaling regime. We have proposed (Oono and Puri 1987) cell-dynamical system (CDS) models of spinodal decomposition, which are highly efficient in terms of computer time usage. These models impose the space-time coarsening, implicit in conventional phenomenological models (e.g. the CHC equation) by using a discretised space-time lattice, which can be considered the result of coarse graining the microscopic model. We believe our models are in the same universality class as the CHC equation. Although we cannot derive our models by conventional discretisation of the CHC equation, we have proposed a new discretisation scheme which results in our models starting from the CHC equation†. Our model for the conserved order parameter case is

$$\psi(t+1, n) = \mathcal{F}[\psi(t, n)] - \langle\langle \mathcal{F}[\psi(t, n)] - \psi(t, n) \rangle\rangle \tag{2}$$

where

$$\mathcal{F}[\psi(t, n)] \equiv f(\psi(t, n)) + D(\langle\langle \psi(t, n) \rangle\rangle - \psi(t, n)). \tag{3}$$

In (2), $\psi(t, n)$ is the value of the order parameter in the cell n at time t, D is a positive constant and f is an injection on R (real number line) with two hyperbolic sinks and one hyperbolic source. The sinks correspond to the two new ordered states after quenching. The source corresponds to the single disordered state before quenching. The exact form of f is not crucial. We believe that any map f with properties as described above gives results in the same universality class (Oono and Puri 1988a). We choose $f(x) = A \tanh x$ and $D = 0.5$. In Ooni and Puri (1987), we presented results for $A = 1.3$. In (3), $\langle\langle * \rangle\rangle - *$ is the isotropised discrete Laplacian. We use the following definition of $\langle\langle * \rangle\rangle$ on the square lattice:

$$\langle\langle \psi(t, n) \rangle\rangle = \tfrac{1}{6} \sum (\psi \text{ in the nearest-neighbour cells}) + \tfrac{1}{12} \sum (\psi \text{ in the next-nearest-neighbour cells}). \tag{4}$$

The model defined by (2) and (3) is deterministic. To include the effects of noise we use an analogy to the CHC equation where we can use for the noise

$$\sigma(\boldsymbol{r}, t) = \nabla \cdot \boldsymbol{\eta}(\boldsymbol{r}, t) \tag{5}$$

† One can also arrive at a discrete space-time model by conventional discretisation of a partial differential equation like the CHC equation. If the time increment is small, the modelling is accurate but too many updates are required before the scaling regime (if any) is reached. However, if the time increment is large, then the single-cell dynamics becomes oscillatory or even chaotic (Yamaguti and Hatano 1979) whereas we expect it to be purely relaxational. Thus, it is not numerically efficient to use the usual discretisation of the CHC equation to model spinodal decomposition for long times.

where $\boldsymbol{\eta}(\boldsymbol{r}, t)$ is a vector white noise with Gaussian components which satisfy the usual fluctuation-dissipation relation

$$\langle \eta_i(\boldsymbol{r}, t)\eta_j(\boldsymbol{r}', t')\rangle = \delta_{ij}\delta(\boldsymbol{r}-\boldsymbol{r}')\delta(t-t'). \tag{6}$$

Thus, in our discrete space-time model we can include noise as follows:

$$\psi(t+1, n) = \mathcal{F}[\psi(t, n)] - \langle\!\langle \mathcal{F}[\psi(t, n)] - \psi(t, n)\rangle\!\rangle + B[\eta_x(t, n_x+1, n_y) - \eta_x(t, n_x, n_y) + \eta_y(t, n_x, n_y+1) - \eta_y(t, n_x, n_y)] \tag{7}$$

where B (the noise amplitude) is a third parameter in our model (the others being D and the parameter A in f)†. The noise vector (η_x, η_y) (t, n) consists of two random numbers (uniformly distributed in $[-1, 1]$) assigned at each time t to each lattice site $(\equiv (n_x, n_y))$. We have also performed simulations in which noise has a Gaussian distribution and this made no difference to our results. Furthermore, we have also studied the evolution patterns in the case where we started off with a zero (or non-zero) amplitude of noise and switched on (or off) the noise after a certain number of iterations. The patterns rapidly (within about 10 iterations) went to the noisy (or noiseless) forms we demonstrate below. This indicates that there is no cumulative effect of noise. Thus, any reasonable distribution of noise should give the same results.

We have performed simulations on a two-dimensional lattice of size 100×100 with periodic boundary conditions. Here we report results of representative simulations with $f(x) = 1.3 \tanh x$, $D = 0.5$ and $B = 0.0$ (noiseless case) or 0.3, 0.5 (noisy case)‡. We have actually used a variety of functions f and different values of D, B. Detailed results will be presented elsewhere (Puri and Oono 1988).

All calculations presented here were done on a VAX-750 computer (without using array processors). One update of the lattice in the noiseless case took 1.77 s of CPU time. The corresponding CPU time for the noisy case was 2.98 s. Form factors were calculated as averages over 20 different initial configurations. This was sufficient to get smooth master functions. (An independent confirmation of our results has been obtained by Yeung who calculated statistical averages over 30 different initial configurations. Notice that the statistical error is only halved even if we use as many as 80 initial configurations in our statistics.) The raw data for the form factor are defined as a function of vector $\boldsymbol{k}$ rather than scalar k. Notice that $\boldsymbol{k}$ can take up values (for a lattice of size $N \times N$) $2\pi(m_x, m_y)/N$, where m_x and m_y have integer values from $-N/2$ to $(N/2)-1$. To scalarise our data for the form factor, we average it over shells of inner radius $(n-1)\delta k$ (with $\delta k = (2\pi/N) \times 0.5$, N being the lattice size) and outer radius $n\delta k$ (where n is an integer) in the Brillouin zone. The scalar function thus obtained is what we term $S(k, t)$ and the corresponding k value is assigned as

† If we regard our CDS models as arising from a discretisation (albeit an unconventional one) of the corresponding partial differential equations, the noise amplitude B is related to the kinetic coefficient and the discretisation mesh size through the fluctuation-dissipation theorem. Thus, changing B would correspond to changing the kinetic coefficient, if we keep the mesh size fixed. However, in the usual interpretation of the deterministic CHC equation we simply discard the noise while retaining the usual kinetic coefficient. Thus our comparison in this letter is between the noisy case (which does satisfy the fluctuation-dissipation theorem) and the deterministic case (which does not satisfy the fluctuation-dissipation theorem). It should be noted, furthermore, that irrespective of the kinetic coefficient we would expect the asymptotic behaviours to be universal. This is borne out by results we present in a forthcoming paper.

‡ We do not discuss here relations between parameters in our models and those in other (Monte Carlo, Langevin) models. However, we note that results shown here correspond to the case of deep quenching.

$(n-1)\delta k + \delta k/2$ (except in the case $n = 0$, where the corresponding k value is assigned as 0).

In figure 1 we show the patterns obtained from the same inital conditions (namely, randomly distributed values of the order parameter in the range ± 0.125) for the noiseless and noisy ($B = 0.3$) cases. For the noiseless case (shown in the upper frames) the boundary walls are smooth and regular. In the noisy ($B = 0.3$) case (shown in the lower frames) the pattern size at comparable times is of the same order as in the noiseless case, but the boundary walls are ragged. In figure 2 we show a typical pattern for the case $B = 0.5$ with the same initial conditions as before. This pattern is more ragged than the pattern for the case $B = 0.3$ and is similar to previously published patterns from Monte Carlo simulations (Gawlinski *et al* 1985, Huse 1986).

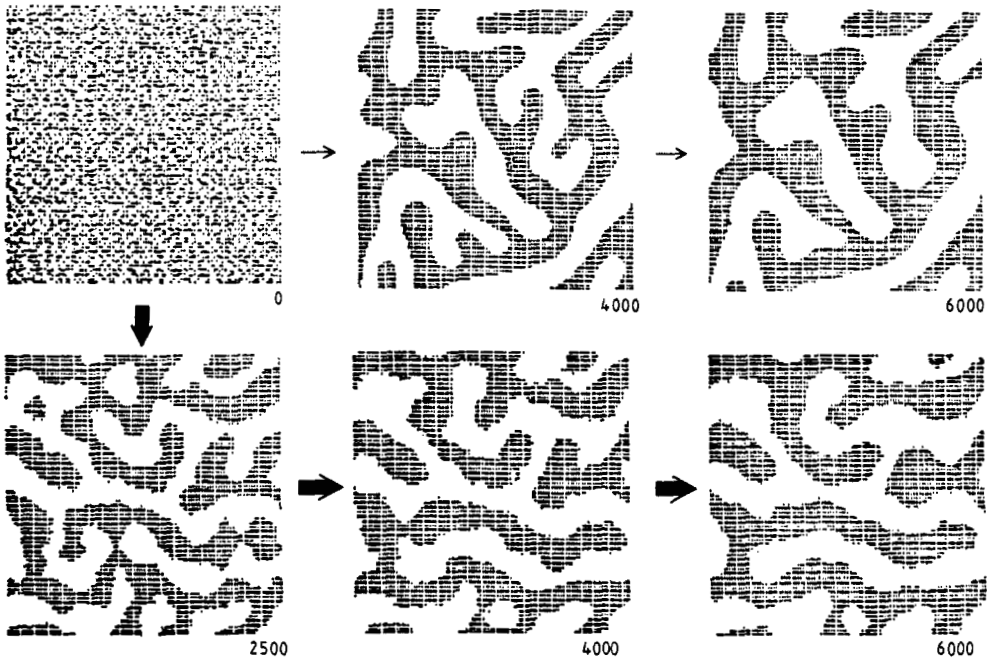

Figure 1. Thin arrows indicate time evolution of patterns for the noiseless case and thick arrows that of the noisy ($B = 0.3$) case, both from the same initial random configuration (labelled 0). The numbers denote necessary time steps from 0. Only points with positive order parameter are marked.

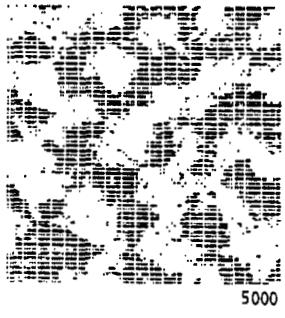

Figure 2. Typical pattern for the strongly noisy ($B = 0.5$) case after 5000 updates.

In figure 3 we show the dependence of $\langle k\rangle(t)$ on time for the noiseless and the noisy cases:

$$\langle k\rangle(t) = \int_0^\infty \mathrm{d}k\, kS(k,t)\left(\int_0^\infty \mathrm{d}k\, S(k,t)\right)^{-1}. \tag{8}$$

This is related to the typical domain size as $\langle k\rangle(t) \propto l(t)^{-1}$. Thus we would expect that (if scaling holds good)

$$\langle k\rangle(t) \propto t^{-\phi}. \tag{9}$$

Numerically, we compute $\langle k\rangle(t)$ by considering all k values up to half the reciprocal lattice size. Figure 3(a) shows $\ln\langle k\rangle(t)$ against $\ln t$ for the noiseless case. The curve

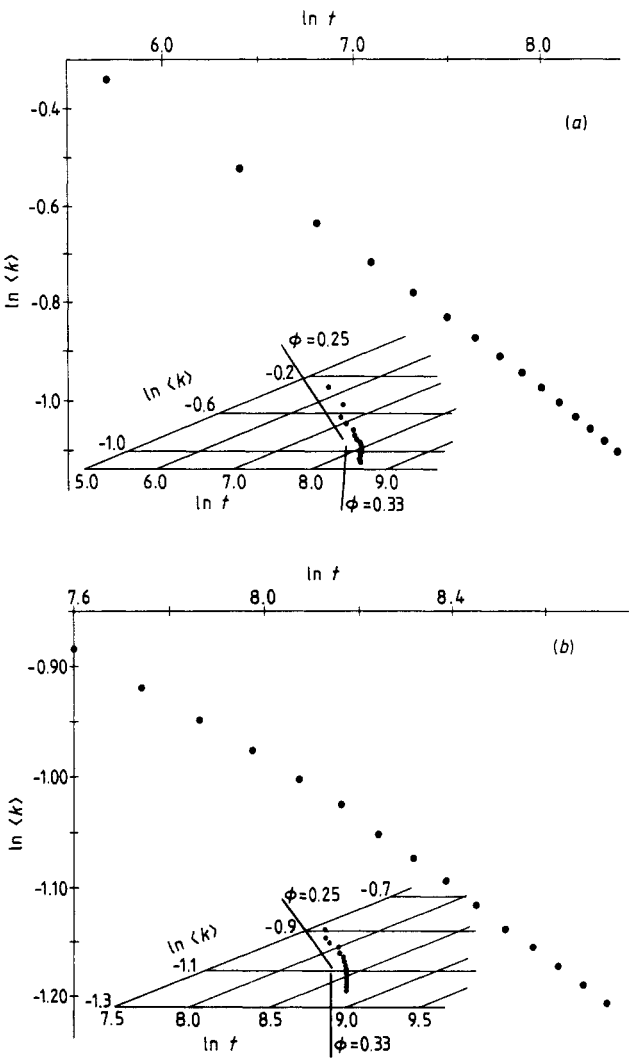

Figure 3. Dependence of $\ln\langle k\rangle$ on $\ln t$ for (a) the noiseless case and (b) the noisy ($B = 0.3$) case. The inset figure shows the data affinely transformed so as to clearly exhibit the crossover behaviour.

shows two linear portions indicating that there is a power-law domain growth but values of ϕ are different in the two regimes. The inset figure shows the data affinely transformed so as to clearly exhibit the crossover behaviour. Essentially, the affine transform used here gives the visual effect of holding the graph almost horizontally at eye level, and then looking along the line of points. We prefer this method to putting straight lines (which may cause prejudice in the observer) through the data points. The slope of the two portions gives the exponent ϕ. There is an apparent crossover at $t \sim 2750$ iterations from an exponent† of $\phi \sim 0.28$ to $\phi \sim 0.33$ (curvature-dominated growth). The earlier time exponent $\phi \sim 0.28$ may suggest a surface diffusion mechanism (see Furukawa's (1985) review article which gives $\phi = 0.25$). Figure 3(b) shows $\ln\langle k\rangle(t)$ against $\ln t$ for the noisy ($B = 0.3$) case. Here, the crossover is delayed to about 3700 iterations. Initially the exponent is $\phi \sim 0.27$ and then crosses over to $\phi \sim 0.33$. Recently, exactly the same model without noise was studied extensively by Chakrabarti and Gunton (1987); they gave $\phi \sim 0.33$ for more than two decades.

In figure 4 we have plotted the scaled scattering function $S(k, t)(\langle k\rangle(t))^2$ as a function of $k/\langle k\rangle(t)$ for different times. In the scaling regime this should be a universal curve. Figure 4 shows data from times 1800, 2400, 3000, 3600 and 4200 for the noiseless case (denoted by circles). They can be seen to lie on a smooth master curve. Notice that data are chosen from either side of the exponent crossover time; the master nature of the curve is *insensitive* to the value of the growth exponent at that time‡. The points marked by crosses in figure 4 correspond to the noisy ($B = 0.3$) case. The data for the

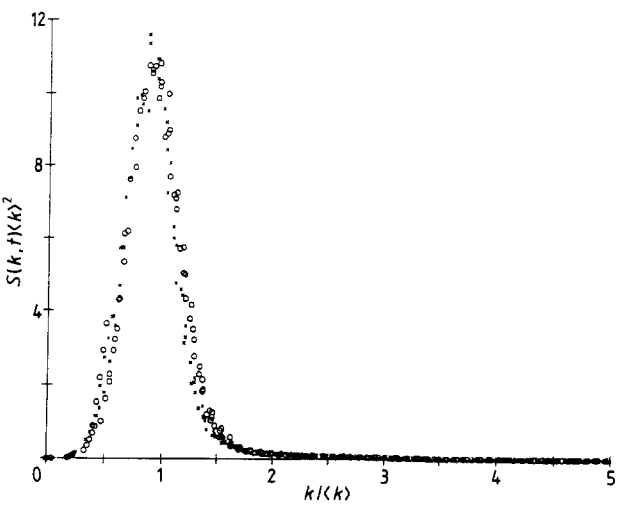

Figure 4. Scaled scattering function $S(k, t)(\langle k\rangle(t))^2$ as a function of $k/\langle k\rangle(t)$ for the noiseless and noisy ($B = 0.3$) cases. The circles are data from the noiseless case at times 1800, 2400, 3000, 3600 and 4200 (through the crossover). The crosses are data from the noisy case at times 2000, 2900, 3800, 4700 and 5600 (also through the crossover). Though it is not evident on the scale of this figure, the tails of the curves do not yet lie on a master curve.

† Exponent values have possible errors of ±0.01. This error is estimated by calculating the slope of extreme lines which can be put through the points under consideration.

‡ Even for much earlier times we can achieve a reasonable master curve by this kind of superposition (Oono and Puri 1988a). However, very long times cannot be fitted to this master curve.

noisy case are for times 2000, 2900, 3800, 4700 and 5600. Again, the data presented for the noisy case are also from both sides of the exponent crossover time. Notice that, in these time regimes, both the noiseless and noisy cases have the same master curve for moderate values of $k/\langle k\rangle(t)$. However, the tails of the curves, which correspond to relatively (compared to pattern size) short wavelength fluctuations, are quite different though this difference is not evident on the scale of figure 4. Porod's law (Porod 1983) is not satisfied for either of the cases. This is a result of the non-zero thickness of the walls, as we discuss elsewhere (Oono and Puri 1988b). For the noiseless case, the tail of the curve drops off faster than x^{-3}, where $x = k/\langle k\rangle(t)$. There is a 'fake Porod's law' in that the data in the extreme tail flatten out. However, this is the result of lattice discretisation. For the noisy case, the initial decay of the tail is faster than x^{-3} as expected. However, because of noise-induced raggedness on very small length scales, the extreme tail of the curve decays slower than x^{-3}, as in the Monte Carlo case.

We can summarise our results as follows.

(A) Irrespective of whether the models are deterministic or stochastic, the long-time behaviours are the same. In particular, we get the same exponent ($\sim$0.33) and expect to get the same master scattering function asymptotically.

(B) For earlier times the exponents are definitely smaller than 0.33 and are closer to 0.25. The crossover time increases with increase in the amplitude of noise.

Since our models can be derived by a special discretisation method from the CHC equation, similar results should be obtained for the long-time behaviour of the CHC equation also.

Our result (A) strongly supports the theoretical consensus (Ohta *et al* 1982) that noise is unimportant in the true scaling regime. Hence we can use our deterministic model to study the asymptotic behaviour of spinodal decomposition. Our result (B) suggests that extant Monte Carlo simulations have not been performed for a sufficiently long time. If we coarse grain a spatial configuration with highly ragged boundary walls, we get a configuration with soft boundary walls, i.e. broad kinks relative to the typical pattern size. In such a configuration, surface diffusion becomes much easier than in the hard-wall case and this results in a smaller exponent ($=0.25$). To verify this we have performed a long-time calculation with $A = 1.2$, which gives rise to softer walls than the case with $A = 1.3$. We find that this results in a delay in the exponent crossover time ($\sim$4000 steps), though the asymptotic exponent is still $\sim$0.33. Thus, what is crucial to the crossover is not the actual size of the pattern but rather the ratio of wall width to the pattern size. Furukawa (1987) has recently argued that the crossover time is proportional to (wall thickness)$^{0.25}$.

We claim that spinodal decomposition modelled by our CDS or by the CHC equation has an asymptotic exponent of 0.33. An exponent smaller than this is a strong indication of insufficient calculation time and/or freezing into metastable states because of the smallness of systems used. In our simulation the observation of the crossover to the asymptotic exponent is enabled by the numerical efficiency of our modelling scheme.

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